

New Real space method for calculation of physical properties of a disordered system.

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We introduce a new real space super cell approximation method for treating the electronic states of disordered systems. This method is general and allows both randomness in the on-site energies and in the hopping integrals. In the special case of randomness in the on-site energies only, this method is equivalent to the Non Local Coherent Potential Approximation (NLCPA) derived previously.

For the past three decades the Coherent Potential Approximation (CPA) was the most widely used self-consistent mean field treatment of disordered systems[1]. As a single site approximation, the CPA Green's functions are averaged over the randomness on a single site, and so could not handle the effects of correlations between the random potentials on neighbouring sites. In order to include such inter site effects extensions of CPA were developed using the locator formalism[2, 3, 4]. However, recently the Dynamical Cluster Approximation (DCA) has been introduced for interacting systems[5]. A version of DCA has also been used to treat disordered systems [6]. This method extends CPA by including a weakly wave vector dependent self energy $\Sigma(k; \omega)$. The DCA was originally derived on basis of restricting the role of momentum conservation in calculating the self-energy, by applying a coarse graining procedure to the First Brillouin Zone (FBZ). Momentum conservation is applied only to the coarse grained wave vector \mathbf{K}_n in the FBZ and in the corresponding Laue function. In this method a key computational step makes use of a Fourier like transformation of coarse grained wave vectors \mathbf{K}_n to a real-space set of coordinates, \mathbf{R}_n . These coordinates were interpreted as the lattice sites of a real cluster of a real lattice, however this connection was not established directly.

By using effective medium theory in real-space we developed a non-local CPA (NLCPA) method. In this method a cluster of impurities is embedded in an effective medium, in such a way that all cluster sites are equivalent and lattice periodicity is maintained for the disorder averaged Green functions. For a one dimensional binary alloy numerically we showed that a periodic super cell density of states is similar to the NLCPA density of states[7]. We also applied NLCPA as a real space super cell approximation to explain the resonance peak that appears in the density of states of substituted Zn impurity in CuO_2 plane of $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+\delta}$ [8]. Subsequently NLCPA was also extended to the case of the first-principles Korringa-Kohn-Rostoker (KKR) CPA model for the electronic structure of disordered systems.[9]

In this paper we show that the super cell self energy periodicity in real space leads naturally to coarse graining in k-space, providing us with an alternative and general

derivation of the NLCPA method. This establishes directly the connection between the real-space cluster and the coarse graining in k-space, showing clearly for the first time that both DCA and NLCPA are equivalent to a super cell approximation in real space.

We start our investigation on a general tight binding model for a non-interacting alloy system,

$$H = - \sum_{ij} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} + \sum_{i\sigma} (\varepsilon_i - \mu) \hat{n}_{i\sigma}. \quad (1)$$

where t_{ij} are the hopping integrals which may be random, μ is the chemical potential and ε_i is the random on site energy which takes values $-\delta/2$ with probability $1 - c$ for host sites, and $\delta/2$ with probability c for impurity sites.

The equation of motion for electrons with the above Hamiltonian, Eq.1, for any general impurity configuration $\{\varepsilon_i\}$ in terms of the Green function is,

$$\sum_l ((\omega - \varepsilon_i + \mu) \delta_{il} + t_{il}) G(l, j; \omega) = \delta_{ij} \quad (2)$$

We expand the full random Green function $G(i, j; \omega)$ in Eq.2, in terms of the clean system Green function $G^0(i, j; \omega)$, [10]

$$G(i, j; \omega) = G^0(i, j; \omega) + \sum_{ll'} G^0(i, l; \omega) V_{ll'} G(l', j; \omega) \quad (3)$$

where the random potential matrix $V_{ll'}$ is

$$V_{ll'} = \varepsilon_l \delta_{ll'} + \delta t_{ll'}, \quad (4)$$

$\delta t_{ll'} = t_{ll'} - t_{ll'}^0$, is the difference between hopping integral in random and clean system and $G^0(i, j; \omega)$ is

$$G^0(i, j; \omega) = \frac{1}{N} \sum_{\mathbf{k}} e^{i\mathbf{k} \cdot \mathbf{r}_{ij}} (\omega - \epsilon_{\mathbf{k}} + \mu)^{-1}. \quad (5)$$

where $\epsilon_{\mathbf{k}} = -\frac{1}{N} \sum_{ij} t_{ij}^0 e^{i\mathbf{k} \cdot \mathbf{r}_{ij}}$ is the band structure for the clean system.

The Dyson equation corresponding to Eq.3 is

$$\bar{G}(i, j; \omega) = G^0(i, j; \omega) + \sum_{ll'} G^0(i, l; \omega) \Sigma(l, l'; \omega) \bar{G}(l', j; \omega) \quad (6)$$

where,

$$\sum_{l'} \langle V_{ll'} G(l', j; \omega) \rangle = \sum_{l'} \Sigma(l, l'; \omega) \bar{G}(l', j; \omega). \quad (7)$$

The disorder average Green function, Eq.6, can be written,

$$\bar{G}(i, j; \omega) = \frac{1}{N} \sum_{\mathbf{k}} e^{i\mathbf{k} \cdot \mathbf{r}_{ij}} (\omega - \epsilon_{\mathbf{k}} + \mu - \Sigma(\mathbf{k}; \omega))^{-1}. \quad (8)$$

where

$$\Sigma(\mathbf{k}; \omega) = \frac{1}{N} \sum_{i,j} e^{-i\mathbf{k} \cdot \mathbf{r}_{ij}} \Sigma(i, j; \omega). \quad (9)$$

is the Fourier transform of the self-energy.

Fourier transformations of this system of equations obey the orthogonality condition,

$$\frac{1}{N} \sum_{\mathbf{k}} e^{-i\mathbf{k} \cdot \mathbf{r}_{ij}} = \delta_{ij} \quad (10)$$

where indices i and j run over all N lattice sites in the crystal. Because the exact solution of Eqs.3 and 6 is impossible, they are usually solved in different single site approximations such as the Born approximation, the T-matrix approximation and the CPA.[7] It is well known that in all these approximations inter site impurity effects has been neglected.

Here we introduce a new approximation, and we show that, for the case of $\delta t_{ll'} = 0$, it is equivalent to the NLCPA [8]. The derivation of the approximation is as follows. Consider a random alloy system that has been divided to N_c super cells. The super cell Green function $G_{sc}(I, J; \omega)$ can be expand as

$$G_{sc}(I, J; \omega) = G^0(I, J; \omega) + \sum_{l, l'} G^0(I, l; \omega) V_{ll'} G_{sc}(l', J; \omega). \quad (11)$$

where I and J refer to sites in the same super cell and l, l' indices refer to the whole lattice. The average Green function corresponding to Eq.11 is,

$$\bar{G}_{sc}(I, J; \omega) = G^0(I, J; \omega) + \sum_{l, l'} G^0(I, l; \omega) \Sigma_{sc}(l, l'; \omega) \bar{G}_{sc}(l', J; \omega). \quad (12)$$

Now we apply an approximation that is based on two assumptions: first *neglect the hopping integrals deviation, $\delta t_{ll'}$, between impurity sites of different super cells, that is, $\delta t_{ll'} = 0$ when l and l' are in different super cells* and second *neglect multiple scattering between impurities in different super cells*. Therefore we have

$$\Sigma_{sc}(i, j; \omega) = 0, \text{ if } i \text{ and } j \notin \text{same super cell}. \quad (13)$$

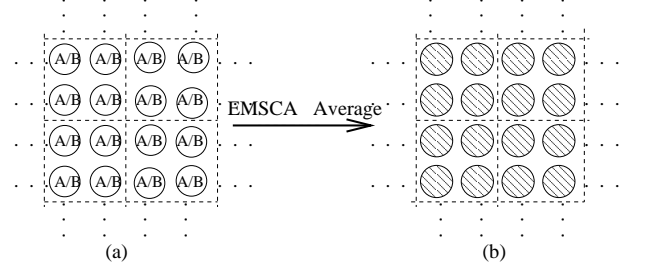


FIG. 1: (a): A binary alloy divided into super cells containing four sites. (b) The approximation of impurity averaging over the super cells. By neglecting the multiple scattering between impurities in different super cells, the self energies in different super cells obey the periodicity law given in Eq.14.

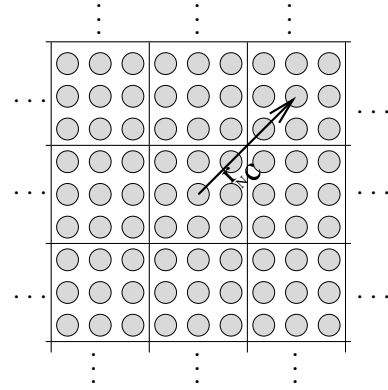


FIG. 2: A two dimensional lattice divided into super cells of nine sites. The arrow indicates the super cell periodicity vector $\mathbf{r}_{N_c} = 3\mathbf{a}_1 + 3\mathbf{a}_2$, where $N_{c1} = 3$ and $N_{c2} = 3$.

Fig.1 shows a binary alloy system divided into periodic super cells containing four impurity sites. By neglecting both the hopping integrals deviations $\delta t_{ll'}$ and also the correlations between different super cells sites, the self energy in each super cell is independent of the other super cells and is periodic with respect to super cell translation vectors. These conditions imply that the disorder averaged system self energy has the following super cell periodicity,

$$\Sigma_{sc}(\mathbf{r}_{IJ} + \mathbf{r}_{N_c}; \omega) = \Sigma_{sc}(\mathbf{r}_{IJ}; \omega) \quad (14)$$

where I and J refer to the sites in the same super cell, and the super cell translation vector \mathbf{r}_{N_c} is

$$\mathbf{r}_{N_c} = \sum_{i=1}^3 m_i N_{ci} \mathbf{a}_i, \quad (15)$$

where $N_c = N_{c1} N_{c2} N_{c3}$ is the number of lattice sites in a 3-dimensional super cell, \mathbf{a}_i are primitive vectors of real lattice, and N_{ci} is the number of sites in the super cell in the i th direction and m_i are integers. Fig.2 shows \mathbf{r}_{N_c} for a two dimensional lattice with periodicity over nine sites in each super cell. Note that when $N_c \rightarrow \infty$

all super cells will coincide and are equal to the full real lattice.

Fourier transformation of Eq.14 implies that we should restrict the self energy wave vectors \mathbf{k} to those obeying

$$e^{i\mathbf{k}\cdot\mathbf{r}_{N_c}} = 1, \quad (16)$$

These are the set

$$\mathbf{K}_n = \sum_{i=1}^3 \frac{l_i}{N_{ci}} \mathbf{b}_i. \quad (17)$$

where \mathbf{b}_i are the reciprocal lattice primitive vectors and l_i are integers. Notice that number of the \mathbf{K}_n in the first Brillouin zone is equal to number of the lattice sites, N_c , in a super cell. Therefore the Fourier transformation of the self energy is

$$\Sigma(\mathbf{K}_n; \omega) = \frac{1}{N} \sum_{i,j} e^{-i\mathbf{K}_n \cdot \mathbf{r}_{ij}} \Sigma_{sc}(i, j; \omega). \quad (18)$$

By inserting Eq.13 into Eq.18 we find that

$$\Sigma(\mathbf{K}_n; \omega) = \frac{1}{N_c} \sum_{I,J} e^{-i\mathbf{K}_n \cdot \mathbf{r}_{IJ}} \Sigma_{sc}(I, J; \omega) \quad (19)$$

where I and J refer to the sites in the same super cell. By converting the Eq.19 to real space we find

$$\Sigma_{sc}(I, J; \omega) = \frac{1}{N_c} \sum_{\mathbf{K}_n} e^{-i\mathbf{K}_n \cdot \mathbf{r}_{IJ}} \Sigma(\mathbf{K}_n; \omega) \quad (20)$$

where the orthogonality condition in the super-cell is,

$$\frac{1}{N_c} \sum_{\mathbf{K}_n} e^{-i\mathbf{K}_n \cdot \mathbf{r}_{IJ}} = \delta_{IJ} \quad (21)$$

Because the set of $\{\mathbf{K}_n\}$ vectors divides the volume of the first Brillouin zone to N_c equal patches, we therefore identify each patch, n , by the corresponding wave vector \mathbf{K}_n that is located at its center. Therefore we define the relation between \mathbf{K}_n and \mathbf{k} inside each patch as follows

$$\mathbf{k} = \mathbf{K}_n + \mathbf{k}'_n \quad (22)$$

where \mathbf{k}'_n are the wave vectors inside of n th patch with respect to the center of the patch. Fig.3 illustrates this relationship, Eq.22, for one of the patches in a two dimensional system for $N_c = 9$. By comparing Eqs. 21 and 10 we find that the super cell approximation is equivalent to the replacement

$$e^{i\mathbf{k}'_n \cdot \mathbf{r}_{IJ}} \approx 1. \quad (23)$$

Considering of the Eqs. 23, 21, 19 and Eq. 12 we find that the super cell approximation Green function $\bar{G}_{sp}(I, J; \omega)$ is

$$\bar{G}_{sc}(I, J; \omega) = \frac{1}{N_c} \sum_{\mathbf{K}_n} e^{i\mathbf{K}_n \cdot \mathbf{r}_{IJ}} \bar{G}(\mathbf{K}_n; \omega) \quad (24)$$

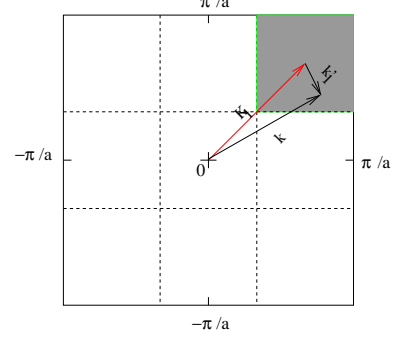


FIG. 3: Relation between \mathbf{k} , \mathbf{K}_n and \mathbf{k}'_n in the First Brillouin Zone (FBZ) for a two dimensional system with a 9 site super cell. As given by Eqs. 17 and 22 there are 9 different \mathbf{K}_n in the FBZ, and so the FBZ is divided into 9 equal areas, while each \mathbf{K}_n is located at the center of one of these areas.

where

$$\bar{G}(\mathbf{K}_n; \omega) = \frac{N}{N_c} \sum_{\mathbf{k}'_n} (\omega - \epsilon_{\mathbf{K}_n + \mathbf{k}'_n} + \mu - \Sigma(\mathbf{K}_n; \omega))^{-1}. \quad (25)$$

In short Eq. 18 and 23 are the super cell approximation conditions. In the limit of $N_c \rightarrow \infty$, $\mathbf{k}'_n \rightarrow 0$ and also $\mathbf{K}_n \rightarrow \mathbf{k}$. Therefore Eqs. 21 and 19 respectively convert to Eqs. 10 and 9 in the exact average system. Also for the exact average system in the limits of super cell periodicity with $N = mN_c$, with m an integer, in the case where correlation between super cells is neglected, the exact average self energy with these conditions equals the super cell periodic self energy, that is

$$\Sigma(I, J; \omega) = \Sigma_{sc}(I, J; \omega). \quad (26)$$

By use of Eq. 23 the Fourier transformation of Eq. 26 can be carried out by reordering the summation over \mathbf{k} as $\sum_{\mathbf{k}} = \sum_{\mathbf{K}_n} \sum_{\mathbf{k}'_n}$ and using the fact that $\sum_{\mathbf{k}'_n} 1 = \frac{N}{N_c}$. We find that

$$\Sigma(\mathbf{K}_n; \omega) = \frac{1}{N_c} \sum_{\mathbf{k}'_n} \Sigma(\mathbf{K}_n + \mathbf{k}'_n; \omega) \quad (27)$$

and Eqs. 10, 9 and 8 will become equal to Eqs. 21, 27 and 24 respectively. This means that the super cell calculation in an effective medium is an approximation for the exact disordered system that includes only the correlation between impurities within a single super cell.

On the other hand, by eliminating the clean Green function matrix \mathbf{G}_0 between Eqs. 3 and 6 the random matrix Green function \mathbf{G} can be expressed in terms of the average Green function $\bar{\mathbf{G}}$ as

$$\mathbf{G} = \bar{\mathbf{G}} + \bar{\mathbf{G}}(\mathbf{V} - \Sigma)\bar{\mathbf{G}}. \quad (28)$$

By applying the super cell approximation with Eq. 13, and taking the average over impurities in all super cells

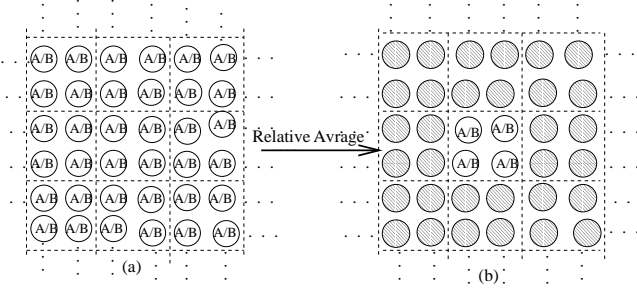


FIG. 4: A four site impurity super cell in an effective medium of mean-field super cells.

except one, Eq. 28 reduces to the following equation for the remaining super cell,

$$\mathbf{G}_{sc}^{imp} = \bar{\mathbf{G}}_{sc} + \bar{\mathbf{G}}_{sc}(\mathbf{V}_{sc} - \Sigma_{sc})\mathbf{G}_{sc}^{imp}. \quad (29)$$

where all the matrices in Eq. 29 are of dimension $N_c \times N_c$. By rearranging Eq. 29 we have

$$\mathbf{G}_{sc}^{imp^{-1}} + \mathbf{V}_{sc} = \bar{\mathbf{G}}_{sc}^{-1} + \Sigma_{sc}. \quad (30)$$

Defining each side of Eq. 30 as a cavity super cell Green function $\hat{\mathcal{G}}^{-1}$ we have

$$\mathbf{G}_{sc}^{imp^{-1}} + \mathbf{V}_{sc} = \hat{\mathcal{G}}^{-1}. \quad (31)$$

and

$$\bar{\mathbf{G}}_{sc}^{-1} + \Sigma_{sc} = \hat{\mathcal{G}}^{-1}. \quad (32)$$

Fig. 4 shows an example four site super cell approximation in which the average has been taken over all super cells except one *impurity super cell*. The matrix elements of Eq. 31 are

$$\begin{aligned} G_{sc}^{imp}(I, J; \omega) &= \mathcal{G}(I, J; \omega) \\ &+ \sum_{L, L'} \mathcal{G}(I, L; \omega) V_{LL'} G_{sc}^{imp}(L', J; \omega). \end{aligned} \quad (33)$$

where the cavity Green function $\mathcal{G}(I, J; \omega)$ obeys

$$\mathcal{G}(I, J; \omega) = \frac{1}{N_c} \sum_{\mathbf{K}_n} e^{i\mathbf{K}_n \cdot \mathbf{r}_{IJ}} \mathcal{G}(\mathbf{K}_n; \omega). \quad (34)$$

Similarly, the matrix elements corresponding to Eq. 32 are,

$$\begin{aligned} \bar{G}_{sc}(I, J; \omega) &= \mathcal{G}(I, J; \omega) \\ &+ \sum_{L, L'} \mathcal{G}(I, L; \omega) \Sigma_{sc}(L, L'; \omega) \bar{G}_{sc}(L', J; \omega). \end{aligned} \quad (35)$$

where the average of the impurity Green function $G_{sc}^{imp}(I, J; \omega)$ over all impurity configurations $\{V_{IJ}\}$ in the super cell is

$$\langle G_{sc}^{imp}(I, J; \omega) \rangle = \bar{G}_{sc}(I, J; \omega). \quad (36)$$

Eqs. 25, 27, 35, 36 constitute a closed set to be solved self consistently.

The above system of equations can be implemented numerically by the following algorithm:

- 1- make a guess for $\Sigma(\mathbf{K}_n; \omega)$, usually zero.
- 2- Calculate $\bar{G}(\mathbf{K}_n; \omega)$ from Eq. 25,
- 3- Use Eq. 35 to calculate the Fourier transform of the cavity Green function

$$\mathcal{G}(\mathbf{K}_n; \omega) = (\bar{G}^{-1}(\mathbf{K}_n; \omega) + \Sigma_{sp}(\mathbf{K}_n; \omega))^{-1} \quad (37)$$

- 4- Calculate the impurity super cell Green function $G_{sp}^{imp}(I, J; \omega)$ from Eq. 33.

- 5- Calculate the average Green function from Eq. 36 and Fourier transform it to \mathbf{K}_n space by using Eq. 21.

- 6- Using Eq. 37 calculate the new self energies $\Sigma_{sc}(\mathbf{K}_n; \omega)$ and go back to step 1 and repeat whole process until convergence has been obtained to a desired accuracy.

As an application of this method we calculate the density of states of a two dimensional square lattice at half band filling, $\bar{n} = 1$ and $\delta = 6t^0$ in which $N_c = 1$ (CPA) and $N_c = 9$. For the $N_c = 9$ case, we also show the effects of introducing random hopping parameters, by considering the two cases, $\delta t_{AA} = 0$, $\delta t_{AB} = 0$ and $\delta t_{AA} = 4t^0$, $\delta t_{AB} = t^0$, where

$$\delta t_{\langle ij \rangle} = t_{AA} - t^0 \quad (38)$$

which i and j are nearest neighbour sites and both are A type, and we also defined,

$$\delta t_{\langle ij \rangle} = t_{AB} - t^0 \quad (39)$$

where at the nearest neighbours sites i and j one is A and another is B type atom. Fig. 5 shows that for the $N_c = 1$ (CPA) and the case of $N_c = 9$ without random hopping, in which $\delta = 6t^0$, we are at the band splitting regime[11, 12]. In this case at half band filling a metal-insulator phase transition is taken place, in spite of different gap sizes. While for the case of $N_c = 9$ with random hopping $\delta t_{AA} = 4t^0$, $\delta t_{AB} = t^0$, the band splitting is not happened, thus the system is a metal. Therefore due to including randomness in both the on-site energies and also the hopping integrals, EMSCA technique can be provide a more realistic results.

Finally, we summarize our discussion with the following conclusions. We have introduced a new real space effective medium super cell approximation (EMSCA) for random systems. For the case $N_c = 1$ this recovers the single site CPA formalism. However for larger values of N_c the method extends CPA by including both the effects of multiple scattering, and by allowing randomness in the hopping integrals. Furthermore, in the limit of $N_c \rightarrow \infty$ the method is exact. The EMSCA in the special case where hopping randomness is neglected, $\delta t = 0$, leads to an alternative derivation of the NLCPA technique for disordered systems. This derivation completely

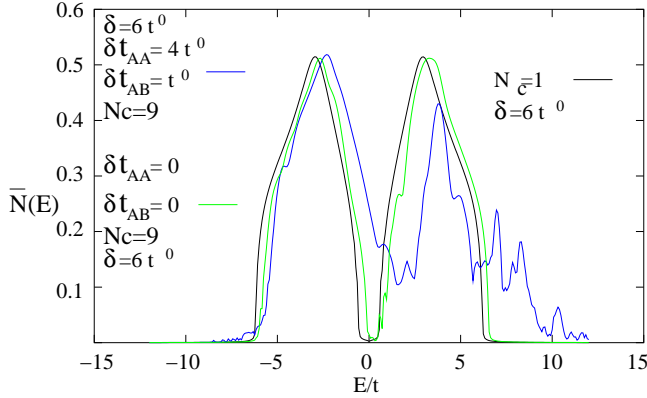


FIG. 5: The density of states of a two dimensional square lattice of a binary alloy A_cB_{1-c} at half band filling, $\bar{n} = 1$, for $c = 0.5$ and $\delta = 6t^0$ when $N_c = 1$ (CPA) and $N_c=9$. We compared $N_c = 1$ DOS and of two cases of $N_c = 9$, first $\delta t_{AA} = 0$, $\delta t_{AB} = 0$ and second $\delta t_{AA} = 4t^0$, $\delta t_{AB} = t^0$. For both non hopping integral randomness cases the band splitting happened (metal insulator phase transition take placed), while for the other not (still is a metal).

establishes the NLCPA as a valid and useful extension of the old and popular CPA method, which incorporates the effects of inter-site correlations.

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